

Terahertz Spectroscopy in the Lab and at Telescopes

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Abstract. The section of the electromagnetic spectrum extending roughly from wavelengths of 3 mm to 30 μm is commonly known as the far-infrared or TeraHertz (THz) region. It contains the great majority of the photons emitted by the universe, and THz observations of molecules and dust are able penetrate deeply into molecular clouds, thus revealing the full history of star and planet formation. Accordingly, the upcoming deployments of the Herschel, ALMA, and SOFIA observatories promise to revolutionize our understanding of THz astrophysics. To fully realize this promise, however, it is essential that we achieve a quantitative experimental understanding of the dust, ice, and gas which make up the ISM. After outlining the tremendous impact that Tom Phillips has had on astronomical applications of THz radiation, this contribution will describe how emerging technologies in ultrafast lasers are enabling the development of integrated frequency- and time-domain THz facilities that can acquire high dynamic range optical constants of the major components that comprise astrophysical dust, ice and organics across the full wavelength region accessible to Herschel and other THz observatories.

1 Introduction

Spectroscopic tools form an essential core of modern astrophysics. The section of the electromagnetic spectrum extending roughly from $0.1\text{--}10 \times 10^{12}$ Hz ($3\text{--}300\text{ cm}^{-1}$) is commonly known as the far-infrared (FIR), submillimeter, or terahertz (THz) window, and therefore lies between the microwave ($\lambda \sim 30\text{ cm} \rightarrow 3\text{ mm}$) and infrared regions. Accordingly, the THz window shares both scientific and technological characteristics with its longer and shorter wavelength neighbors. Tom Phillips has pioneered both the THz detector technology and the scientific application of THz radiation to astrophysical regimes, where the extinction and re-radiation of the optical/UV output of stars by dust renders many of the tracers regarding the evolution and fate of the universe accessible only in the IR and beyond. More generally, imaging and spectroscopy at THz frequencies holds the key to our ability to remotely sense environments as diverse as primeval galaxies, star and planet-forming circumstellar disks, comets, laboratory plasmas, semiconductor circuitry, and hydrogen bonded liquids/polymers.

Being one of Tom's early Ph.D. students, I would like to share a few of the things I have learned from him as a mentor before discussing recent applications of THz spectroscopy to studies of the chemistry associated with the star and planet formation. In no particular order, they are: (1) When building and deploying instruments, know your signal path. This was drilled into me while bouncing around the sky in the Kuiper Airborne Observatory, but it has saved

countless observing runs since and many, many hours in the lab. (2) Here I'll paraphrase Frank De Lucia — "Give yourself a chance to be lucky." This sage advice has been handed down by countless advisors but it is important to take to heart. The Universe is full of surprises. Go find them. (3) Think deeply about what is possible. Prof. Townes's version of this advice is to work in areas that are uncrowded. To do this well, you have to exercise superb scientific judgement, and so it is essential to think about problems/areas of research at their most basic levels. (4) Have a long term plan. It may take a great deal of development to reach the fundamental limits of telescopes or other complex instrumentation. Short term forcings can alter your career both for good and ill, and so having a long term vision is vital for making significant contributions. (5) Never let observing or a meeting ruin a good day on the links. Whether it be golf or another activity, find something that you are passionate about and that helps you to get back to the daily business of science refreshed.

2 THz Astronomy & Star/Planet Formation

In the dense interstellar medium (ISM) characteristic of sites of star formation, scattering of visible/UV light by dust grains renders molecular clouds optically opaque and lowers their internal temperature to only a few tens of Kelvin (before stars are born). The thermal radiation from such objects peaks in the THz region, and only becomes optically thin at even longer wavelengths (Phillips & Keene 1992). The rotational motions of small molecules, the rovibrational transitions of larger species, the softest vibrational/torsional modes of ices, and the phonon modes of solids thus provide, in many cases, the only or the most powerful probes of the dense, cold gas and dust of the ISM. Over the course of Tom's career and largely due to his efforts, ever more sensitive and capable instruments have come into operation that can address the complex chemistry and physics that drives the formation of solar systems.

2.1 The Early Years at OVRO: Massive Star Formation

Strong (sub)millimeter-wave emission from simple molecules such as OH, water, HCN, and especially CO had been used for over a decade to probe the dense ISM when I arrived at Caltech in 1981; and the detection of ever more complex species in targeted searches toward regions of high mass star formation (and long carbon chains toward TMC-1 and in carbon-rich circumstellar shells) was a regular occurrence. Motivated by the rich spectra of stars, an unbiased survey was undertaken toward the Orion KL hot core as part of my thesis using the first 230 GHz Superconductor-Insulator-Superconductor (SIS) receiver deployed on antenna #1 at the Owens Valley Radio Observatory Millimeter Array.

An overview of the results is shown in Figure 1. We had previously put out sections of the spectra on the floor of the control building at OVRO, but the idea to create a single, compressed plot was Tom's; and it strikingly conveys the richness of the mm-wave spectra of hot cores such as Orion. Indeed, lines blanket so much of the frequency space that the integrated emission from molecules is a significant fraction of the overall mm-wave luminosity of the hot core (Sutton et al. 1984). The excitation constraints from such a survey are stringent, and the molecular identifications rather more secure than is possible with selected

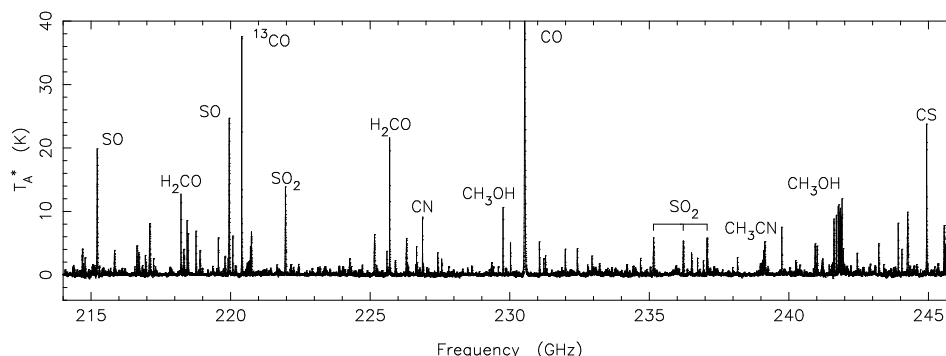


Figure 1. The OVRO 1.3 mm spectral line survey of Orion KL (adapted from Blake et al. 1987). A survey RMS of ~ 0.2 K was achieved after 20+ nights of integration.

integrations over limited regions. Indeed, for a chemist like me it was sobering to realize the number of species detected in the ISM actually dropped slightly as a result of this survey.

2.2 Sun-like Protostars and the CSO

Young, massive stars greatly impact the surrounding gas and dust. Thus, a long standing question concerned whether or not the chemistry in the vicinity of Sun-like protostars showed a similar complexity to that of Orion KL. With the erection of the Caltech Submillimeter Observatory (CSO) atop Mauna Kea and the ever-improving capabilities of SIS receivers, in the 1990s it became possible to acquire very deep integrations of low-mass protostars along with ‘mini-surveys’ in the 230/345 GHz atmospheric windows that assessed the overall level of emission away from the bright lines of simple molecules.

Toward sources like NGC 1333 IRAS 4 and IRAS 16293-2422 molecular depletion was found to be rather more important in the outer reaches of the dense, cold envelope (Blake et al. 1995; van Dishoeck et al. 1995). In the very innermost reaches where the dust is warmed by the central protostars elevated abundances of the critical organics methanol and formaldehyde were seen at levels that could not be created by gas phase chemical networks (Schöier et al. 2002) given the very short transit time of infalling material across the inner regions of the collapsing envelope. Speculations that grain mantle chemistry was critical to the molecular complexity of hot cores was then spectacularly confirmed by very deep searches with the IRAM 30 m and JCMT toward these sources that detected the complex species dimethyl ether and methyl formate (Cazaux et al. 2003).

The improved site and atmospheric transmission also enabled the extension of the Orion survey to other hot cores and to frequencies nearing the atmospheric cutoff near 1 THz. More recently, with very wideband IF receivers and backends it has been possible to repeat the earlier mm-wave surveys but with ten times better sensitivity (and in 1/10th the time, Widicus Weaver, priv. commun.).

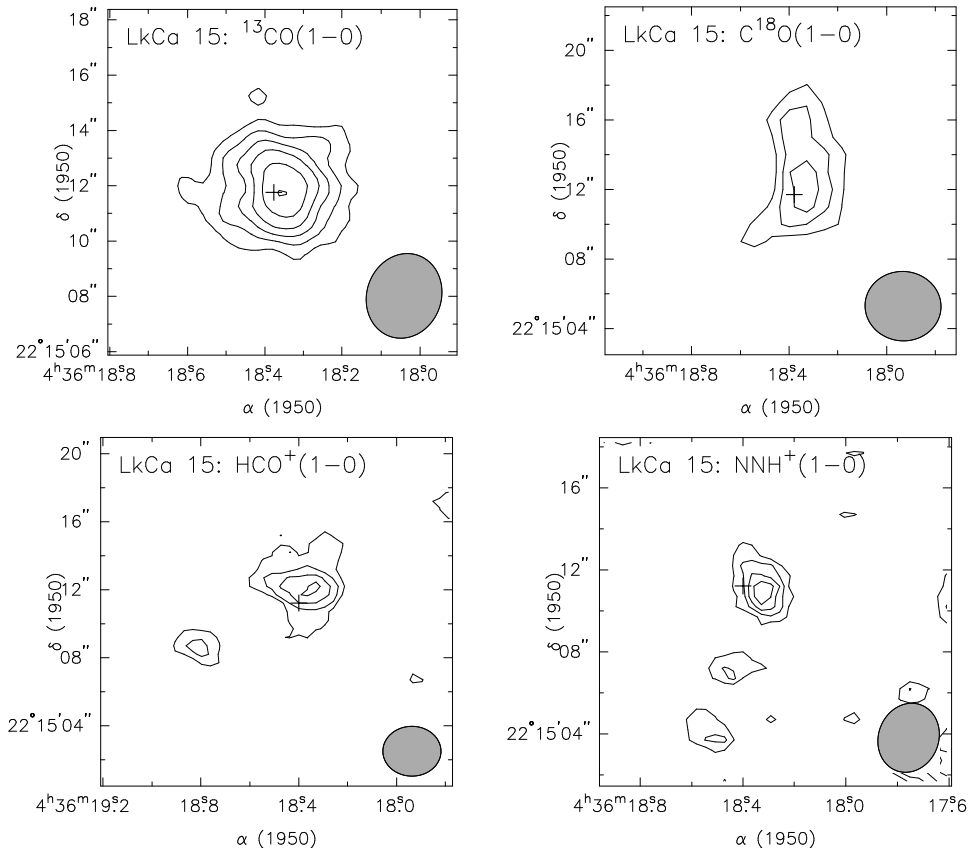


Figure 2. OVRO Millimeter Array interferometric images of the ^{13}CO , C^{18}O , HCO^+ , and N_2H^+ $J = 1-0$ emission from the disk encircling LkCa 15 (Qi et al. 2003). Hatched ellipses at lower right present the synthesized beams. Recent CARMA maps have resolved an inner hole in the mm-dust emission of this source (Carpenter et al., this volume).

2.3 Chemistry in Disks with the OVRO Millimeter Array

Even with the improved sensitivity offered by the CSO and other THz single aperture telescopes, studies of the chemistry in the circumstellar disks in which planet formation occurs is not possible. The amount of gas and dust available within an accretion disk and the timescale over which they are dissipated play major roles in determining what kind of planetary system, if any, can be formed. Furthermore, an understanding of how various volatile species (water, carbon monoxide, methane, ammonia, nitrogen, etc.) are distributed in the outer regions of circumstellar disks is particularly important to examining the connection between interstellar and nebular processes in the formation of icy planetesimals such as comets and Kuiper Belt Objects.

Millimeter-wave interferometric arrays offer a means of studying at least the outer disk; but while a number of systems have been imaged in the most abundant tracer, CO, only a few of the very largest or closest systems have been the subject of chemical studies. One of these, LkCa 15, is presented in

Figure 2. The age, large size, and mass of the LkCa 15 disk make it an important system for further study since it may represent an important transitional phase in which viscous disk spreading and dispersal competes with planetary formation processes (TW Hya, DM Tau, and GM Aur likely present additional examples). The putative inner gap or hole in this source has recently been imaged in $0''.3$ data from CARMA, as is described by Carpenter et al. in this volume. Until the advent of ALMA, the sensitivity of existing arrays is such that only the disk gas beyond $R \sim 40\text{--}50$ AU can be studied (see, for example, Pietu et al. 2007), and so such large systems provide the only opportunity at present to shed light on the chemical and physical gradients in disks.

Ions such as HCO^+ (see Fig. 2) provide critical measurements of the fractional ionization in disks, and thus the likelihood of magnetohydrodynamical sources of viscosity. Another important suite of measurements that can now be undertaken with arrays concerns the constraint of D/H ratios in both ions and neutrals. The first such measurements have recently been made for TW Hya (Qi et al. 2008), and will enable the first direct comparisons with the molecular and isotopic composition of cometary ices.

2.4 An IR Detour: Chemistry in the Planet-Forming Region of Disks

Even with ALMA, it will be difficult to probe the region inside of a few AU in molecular lines. One of the most intriguing questions in the study of the formation of planets, and of terrestrial planets in particular, is how water and volatile organics are transported to their surfaces, and whether or not water is a common ingredient during their early formation and evolution. Observations of water vapor (and water ice) in extrasolar planetary systems, and in proto-planetary disks undergoing planetary formation, at $< \text{AU}$ scales, would be instrumental in resolving these questions.

By combining high dynamic range observations of disks with the Spitzer Space Telescope InfraRed Spectrometer (IRS) with high dispersion echelle observations from 8–10 m class ground based telescopes, we have recently found that intense infrared molecular emission from the near-surface regions of the disks around classical T Tauri stars (cTTs, Salyk et al. 2008; see also Carr & Najita 2008). Sample spectra of AS 205 and DR Tau are shown in Figure 3. Further, using the spectro-astrometric mode of CRIRES at the VLT, we have shown that spatial information at < 0.1 AU scales can be recovered from M-band observations of the $\text{CO } \Delta v=1$ emission in disks (Pontoppidan et al. 2008). Ongoing surveys with Spitzer and Keck/VLT reveal that the emission is common around cTTs, but not Herbig Ae stars (Mandell et al. 2008), and the mid-IR emission from water is found to be strongly sub-thermal (Meijerink et al. 2009).

2.5 The Promise of Herschel, SOFIA, and ALMA

The widespread water vapor emission from disks observed with Spitzer should continue well into the ranges observable with Herschel, SOFIA, and ALMA, as is shown in Figure 4 (Pontoppidan et al. 2009). In particular, the strong lines in the Herschel PACS and HIFI ranges are found to be very sensitive to the amount of residual water vapor in the disk *outside* the snow line (Meijerink et al. 2009); and thus are expected to be highly complementary to the tracers available to Spitzer and ground-based infrared telescopes. Some radial information will be available

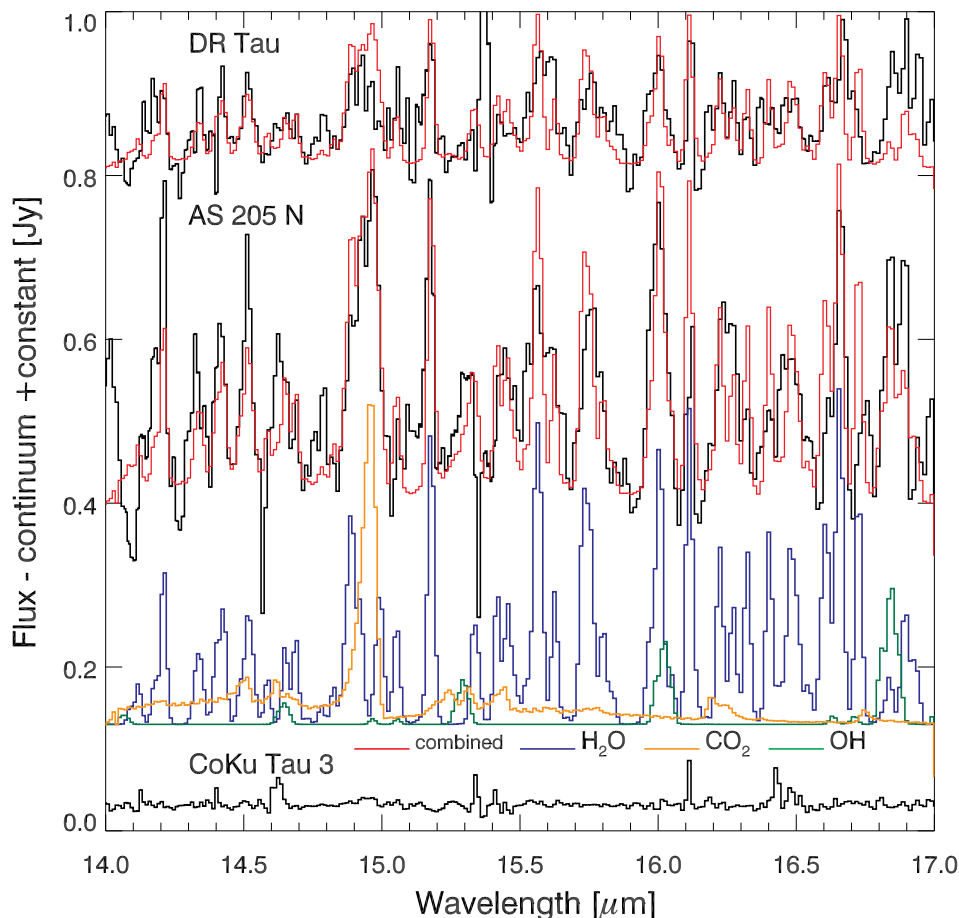


Figure 3. A portion of the Spitzer-IRS spectra of DR Tau and AS 205N (Salyk et al. 2008), compared to LTE models of H_2O , OH, and CO_2 emission. Data are in black. The spectrum of CoKu Tau 3, a source of similar brightness to those presented, is included to demonstrate the dynamic range and the level of systematics such as residuals from the de-fringing process. In all spectra continuum splines have been used to remove the broad emission from silicates.

from the lineshapes as measured by HIFI, but much more direct constraints will be generated by high angular resolution ALMA observations of high excitation H_2^{16}O lines along with more moderate excitation transitions from the oxygen isotopologues and HDO.

More uniquely, perhaps, Herschel is set to offer dramatically new opportunities for the discovery of complex extraterrestrial organics through searches for the lowest frequency torsional or bending vibrations of molecules. With such modes *both* polar and non-polar species may be examined, in either the gas phase or in the solid state, just as in the mid-IR. Furthermore, the spectral line confusion should be eased by the “spreading out” of the low energy vibrations, and complete rotational resolution for systems in excess of 30 atoms in size should

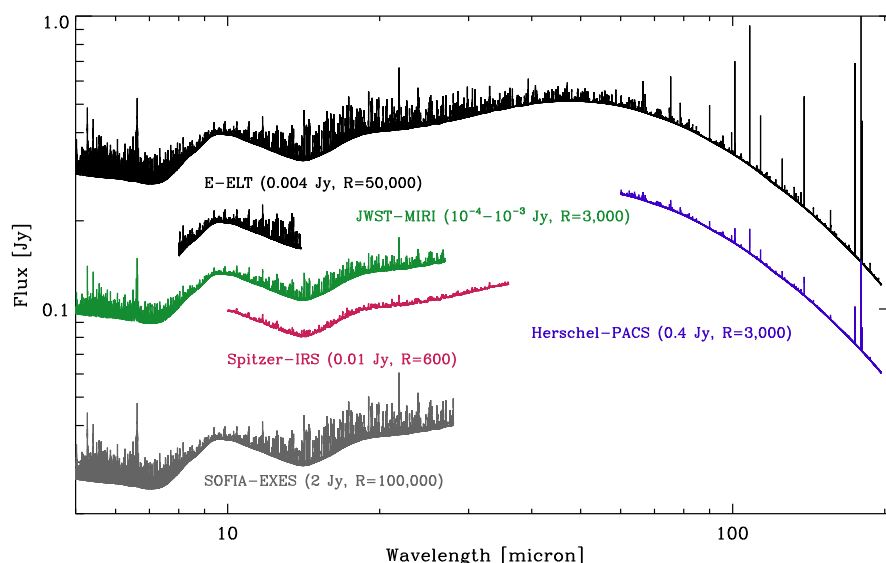


Figure 4. A full range calculation of the infrared water spectrum from a typical protoplanetary disk. In addition, scaled (for clarity) model spectra convolved to the spectral resolution of various infrared observatories are shown. For each observatory and instrument, rough sensitivities ($\sim 10\sigma$ in 1 hour) and spectral resolving powers are indicated. From Pontoppidan et al. (2009).

be readily possible (at least in the lab), since far-IR doppler-limited line widths are typically only 1–10 MHz.

For organics some 20–25 atoms in size with gas phase abundances 10^{-12} that of H_2 , the expected band/continuum ratio for high mass protostars such as Orion or Sgr B2 is several percent. Thus, there is every expectation that new discoveries can be made now that Herschel has been successfully launched and the instruments are performing according to pre-launch expectations. The laboratory challenge should not be underestimated, however, since the generality and uniqueness to molecular structure associated with THz torsions/vibrations comes at a price. These dynamically complex modes are extremely difficult to predict theoretically, and so the spectroscopic search space is very large (unlike that at microwave frequencies). Even if only rotational contours are obtained with Herschel (PACS, for example), it is necessary for laboratory data to ultimately reach full rotational resolution in order to generate synthetic band profiles for any realistic set of ISM conditions. I conclude this short contribution with a few speculations on the kinds of high sensitivity, broad coverage laboratory tools that will aid in the quest to better understand the THz properties of representative interstellar materials.

3 THz Time Domain Spectroscopy of Materials

Even with the rapid scanning speeds of state-of-the-art BWO and multiplier chain LO sources developed for Herschel, it would take several weeks of con-

tinuous scanning to cover the 0.5–3.5 THz range with continuous wave (CW) spectrometers! Complementary tools are therefore needed to rapidly, if coarsely, locate spectral features of interest; and that are equally at home investigating solid, liquid, and gaseous samples. Fortunately, the maturation of femtosecond laser technology has led to THz Time Domain Spectroscopy (THz TDS) techniques which can rapidly accelerate the pace of experimental discovery, as we describe next, and which forms the basis of the research proposed here.

THz pulse generation is an active area of research, with roots that trace back at least three decades. Pulses propagating in free space can be generated either by fast recombination time semiconductors or via the electro-optic (E-O) effect (for excellent overviews of such techniques, see the book edited by Dexheimer 2008). In the first approach, THz antennae are placed onto optoelectronic substrates such as GaAs and illuminated with ultrafast laser optical pulse trains to generate and detect sub-picosecond (ps) THz pulses with optical bandwidths of $\nu \geq 2\text{--}4\text{ THz}$ ($\lambda \leq 80\text{ }\mu\text{m}$). More recently, it has been found that THz pulse detection via the E-O effect in materials such as ZnTe and GaSe offers better sensitivity and larger bandwidths (Schmittenmaer 2004). Importantly, the gated detection of sub-ps pulses via the E-O effect measures the THz electric *field*, and enables both the real and imaginary refractive indices of materials to be measured to high precision. The frequency coverage is analogous to FTS, but since the technique is *coherent* the overall dynamic range can be $>10^6$ with the available powers of 10's of μW , and no cryogenics are required.

The alternative approach to THz pulse generation again involves the E-O effect in materials such as ZnTe. Due to phase matching constraints the generation efficiency at $\sim 800\text{ nm}$ is rather less than that for photoconducting antennas, but the electro-optic approach offers the significant advantage of *extremely* large bandwidths, $> 30\text{ THz}$ under optimum conditions. This approach requires very high peak power, however, well beyond that available with standard Ti:Sapphire ultrafast laser oscillators. Thus, with funds from the NSF Chemistry Research Instrumentation and Facilities: Instrument Development (CRIF:ID) and NASA APRA programs we are in the process of deploying both semiconductor-based and E-O generated THz TDS instruments using state-of-the-art Coherent (Inc.) Ti:Sapphire lasers and amplifiers:

A 1 kHz Regenerative Amplifier 30 fs THz TDS system: The basic layout of the Caltech THz TDS system is shown in Figure 5. The drive laser is a Coherent Legend Elite UltraShort Pulse (USP) system that delivers $< 25\text{ fs}$ pulses of energy $> 3\text{ mJ}$ at a repetition rate of 1 kHz — sufficient energy densities to create intense THz pulses from ZnTe over areas of $\sim 1\text{ cm}^2$. Detection occurs via monitoring of the birefringence induced on a second ZnTe crystal by the THz field using a Wollaston prism and matched 800 nm -sensitive photodiodes. As is true with most THz TDS systems, an opto-mechanical delay line is used to record the THz field as a function of (delay) time. A FFT of the time domain signal yields the spectrum, which after appropriate processing yields both the real and imaginary indices of refraction of the sample under study.

For astrophysical studies it is essential that a wide range of temperatures be achievable, and so we have incorporated into our THz TDS system a cryogens-free, temperature regulated $7\text{--}300\text{ K}$ cold head (Janis Research). The four mirror optical layout permits each of these sample holders to be housed in the purged optical path without the need for (re)alignment, etc. As Figure 5 shows, we can

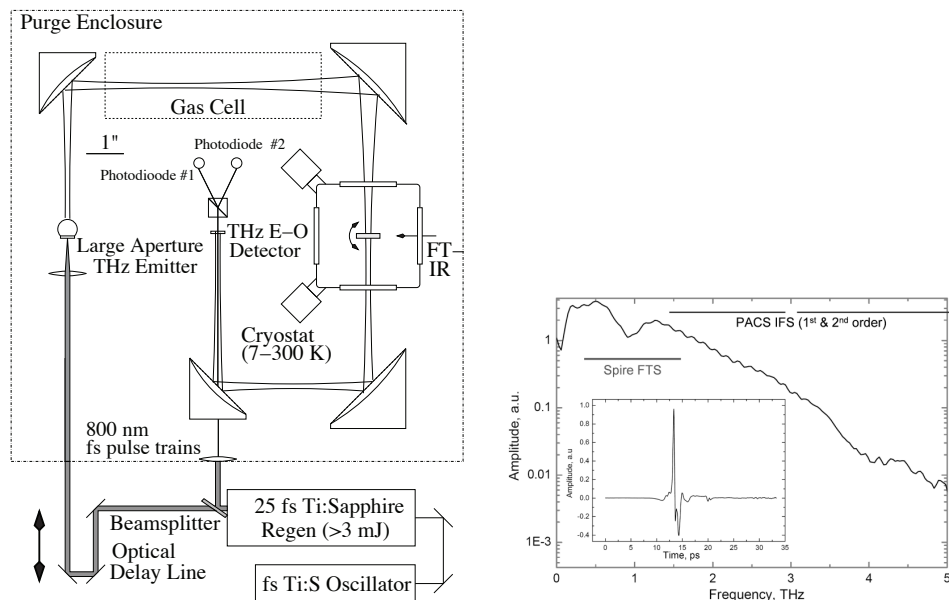


Figure 5. (Left) Schematic diagram of the Caltech Ti:Sapphire regenerative amplifier-driven THz TDS, based on electro-optic generation and sampling in ZnTe and GaSe. The Coherent Legend Elite amplifier provides <25–30 femtosecond 800 nm pulses at 1 kHz (pulse energy >3 mJ) (Right) THz TDS temporal response (inset) and power spectrum for 1 mm thick ZnTe emitters/detectors in a nitrogen-purged atmosphere. The optical delay line can provide a frequency resolution down to $\sim 0.03 \text{ cm}^{-1}$ (1 GHz). Moving to 0.1 mm thick ZnTe will decrease the signal somewhat but extend the spectrometer bandwidth to beyond 10 THz. The SPIRE FTS and PACS IFS spectroscopy ranges are indicated.

therefore acquire THz spectra over the complete PACS and SPIRE ranges with a single instrument, and under conditions relevant to astrophysics.

An all electronic 20 fs THz time domain spectroscopy (TDS) system: As Figure 5 depicts, most existing THz TDSs use optical delay lines to generate the appropriate delays between the optical pulses that gate the THz emitter and receiver. Such opto-mechanical methods, while cost effective and quite useful, suffer from drawbacks in acquisition time and spectral resolution. For example, the scan in Figure 5 took ~ 10 minutes to acquire. Only a fraction of the output of the Ti:Sapphire oscillator used to seed the regenerative amplifier is needed for the high pulse energy THz TDS system, and the remaining light is easily sufficient to drive a second, high repetition rate E-O pulse detection setup.

Thus, to improve upon these limitations, we are implementing a dual laser scanning system in which the relative delay of two mode-locked ultrafast lasers is varied *electronically* by means of active stabilization of their respective cavity lengths (for an extended discussion of such techniques, see Bartels et al. 2007). Very fast averaging times can be achieved with this approach, and the total delay can be easily and optimally adjusted for each experiment out to $1/(\text{rep rate})$ of the oscillators. Accurate and precise calibration of the electronic delay of the pulse trains is critical, however. We use single frequency pump lasers and

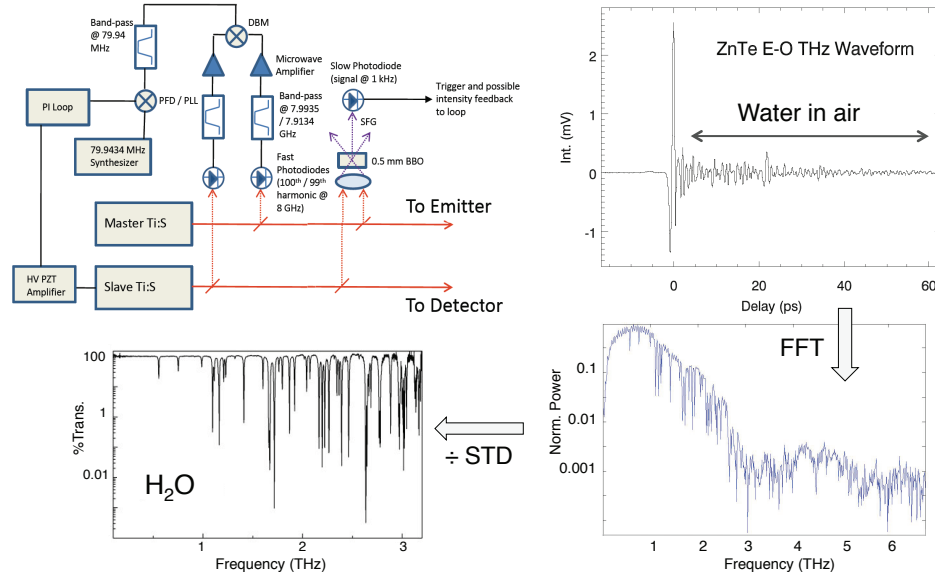


Figure 6. An overview of the dual oscillator asynchronous optical sampling time domain THz spectrometer (top left). The stabilization scheme results in jitter below 100 fs, and absolute frequency calibration to better than a part in 10^{12} . At top right the time domain signature acquired with this instrument in ~ 20 seconds is displayed. The “ringing” after the main pulse is due to atmospheric water vapor, as is shown in the power spectrum produced by an FFT of the time domain data at lower right. When divided by a purged background scan, very high SNR spectra can be acquired in a matter of seconds with a resolution of ~ 100 MHz and no moving parts.

extremely stable Ti:Sapphire laser cavities to achieve such low jitter, and have deployed a dual laser Coherent Mira-5 system that produces pulses of < 20 fs duration at a repetition rate of 80 MHz (average power = 450 mW).

Early results with the new dual oscillator system are shown in Figure 6. The repetition rate of the master laser is locked to an atomic Rb standard, and the slave laser is offset locked to the master using the $\sim 10^{th}$ harmonics near 1 GHz to improve the jitter performance. THz generation uses a commercial large area GaAs-emitter, detection is via the E-O effect in ZnTe. The data in Figure 6 were averaged over 20 seconds. As the FFT and resulting water spectrum show, excellent sensitivity is obtained up to 3 THz, with usable signals up to 6 THz. With high finesse enhancement cavities (Jones et al. 2005) to drive ZnTe E-O emitters at 80 MHz, we expect to generate average THz powers > 1 mW with significant output to $\lambda \geq 30 \mu\text{m}$ — meaning that spectra of the quality depicted in Figure 6 and over the complete PACS and SPIRE ranges will take seconds, not minutes (or hours in the case of incoherent FTS) to acquire.

Acknowledgments. The research described here was largely made possible by the career-long accomplishments of Tom Phillips, to whom I owe an enormous personal and professional debt. I would also like to acknowledge the many students and postdoctoral fellows at Caltech and at Leiden in the group of Prof. Ewine van Dishoeck whom it has been my great privilege to mentor in turn.

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